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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 5 : C08L | A2

(11) International Publication Number: WO 93/19122

(43) International Publication Date: 30 September 1993 (30.09.93)

(21) International Application Number: PCT/US93/02302

(22) International Filing Date: 15 March 1993 (15.03.93)

(30) Priority data:
842,281 17 March 1992 (17.03.92) US

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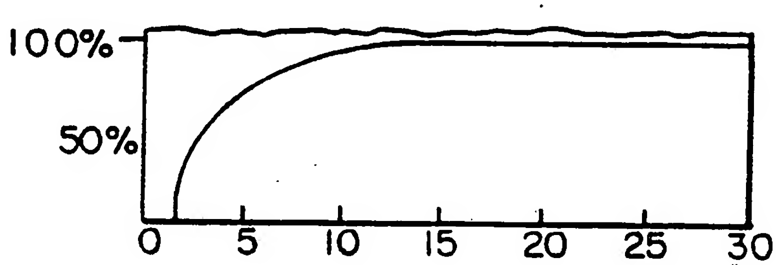
(81) Designated States: JP, KR, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
Without international search report and to be republished upon receipt of that report.

(54) Title: CLEAR HEAT CURABLE SILICONE RUBBER COMPOSITIONS WITH RESIN FOR MATCHING REFRACTIVE INDEX



(57) Abstract

The present invention is based upon the discovery that a relatively small amount of an MQ resin acts as a compatibilizer between components predominantly containing D and Q units respectively. In one embodiment, a vinyl containing organopolysiloxane gum and a silica filler are combined with an MQ or MDQ resin or blends thereof to produce a clear silicone rubber which is heat curable in the presence of an organo peroxide catalyst.

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CLEAR HEAT CURABLE SILICONE RUBBER COMPOSITIONS WITH RESIN FOR MATCHING REFRACTIVE INDEX

BACKGROUND OF THE INVENTION

The present invention relates to heat curable silicone rubber compositions. More particularly, the invention relates to heat curable silicone rubber compositions having improved clarity while maintaining desirable tear strength, oil resistance and compression set properties.

Clear, heat curable silicone rubber compositions are known in the art. One such known composition comprises a mixture of one or more polymers, a high surface area filler of limited particle size and an index matching phenyl containing gum. Blends comprising polydiorganosiloxane gum having 5.3 mole percent phenyl substituents, and a silica filler have been reported to provide substantially clear rubber compounds exhibiting an 83.7 percent transmission, a yellowing index of 4.9, and a haze of 17.1 percent. However, phenyl based compositions are undesirable because they are costly to produce and may release harmful by-products over time. In particular applications, such as medical tubing and devices, non-phenyl based materials are needed which not only have good physical properties but also have a high degree of clarity and low haze.

Additional properties which are of importance in a clear silicone rubber composition are those which are associated with strength. In particular, tear strength, tensile strength, elongation, compression set and hardness are physical properties of interest. Tear strength, measured in pounds per inch (pi), is the ability of the material to withstand continual abrasion without breakdown in mechanical structure. Good tear strength is desirable because it is a measure of toughness. In addition to toughness, elastomeric

materials for most applications should have high tensile strength, measured in pounds per square inch (psi). The tensile strength of a material is related to its ability to resist damaging tensile or pulling forces. At the same time, such materials require good elongation, which is the ability of the material to stretch without breaking. Elongation is measured as a percent of the original sample size. For some applications, hardness, measured on a shore A scale, is an important physical property. Other properties such as resiliency or the ability to recover shape after deformation, especially by compressive forces, may be desirable in a clear rubber product.

Efforts have been undertaken in the past to produce heat curable silicone elastomer compositions having good physical properties. Such efforts are discussed herein by reference to patents set forth below.

U.S. Patent No. 4,539,357 (Bobear) discloses a heat curable silicone composition containing blends of high viscosity, vinyl terminated diorganopolysiloxane gums with a hydride-containing polysiloxane and peroxide curing agent. Although the composition may further comprise a vinyl containing organopolysiloxane resin copolymer of up to about 50 weight percent, Bobear '357 is primarily concerned with tear strength. Clarity of the resulting product is not evaluated among the properties of interest. Similarly, U.S. Patents Nos. 3,652,475 and 3,671,480 to Wada et al. disclose heat curable elastomeric materials. However, clarity is not evaluated among the properties of interest.

U.S. Patent No. 4,061,609 to Bobear discloses a platinum catalyzed silicone rubber composition having a hydride resin component. In the examples, tear

strength is evaluated but clarity is not. U.S. Patent No. 3,660,345 to Bobear shows a further example of an organopolysiloxane elastomer.

U.S. Patents Nos. 3,696,068 to Creamer and 3,884,866 to Jeram et al. also disclose heat curable compositions but clarity is not discussed.

U.S. Patent No. 4,891,393 to Hirai et al. discloses a thermal setting organopolysiloxane composition made from a gum with reinforcing silica and small percentages of methoxy groups, organohydrogenpolysiloxane and a curing catalyst. The materials exhibit improved adhesion to a variety of substrates including metals, glasses and plastics. Some examples listed in Table 4 appear to be transparent.

U.S. Patent No. 4,746,699 to Modic discloses a curable silicone coating composition containing resinous organopolysiloxane components. The materials, which are compounded free of finely divided inorganic fillers, are reported as being transparent whereas compositions containing such fillers are translucent, or opaque depending upon the filler employed.

U.S. Patent No. 4,041,101 to Jeram discloses a solvent resistant silicone rubber composition having a blend of vinyl containing polymers and resins, and a hydride containing resin or hydrogen containing polysiloxane cross-linking agent. A filler may also be present. Clarity does not appear to be discussed.

SUMMARY OF THE INVENTION

The present invention is based upon the discovery that a relatively small amount of an MQ resin acts as a compatibilizer between components predominantly containing D and Q units, respectively. In one embodiment, a vinyl containing organopolysiloxane gum

and a silica filler are combined with an MQ or MDQ resin or blends thereof to produce a clear silicone rubber which is heat curable in the presence of an organo-peroxide catalyst.

The present invention provides clear, heat curable silicone rubber compositions having good strength properties. The compositions comprise, by weight:

(A) about 100 parts by weight of a vinyl containing organopolysiloxane having a viscosity of about 3,000,000 to about 100,000,000 centipoise (cps) at 25°C;

(B) about 1 to about 30 parts by weight of organopolysiloxane resin copolymers having MQ or MDQ units where each M unit can represent M or M' e.g., M-vinyl, units and where each D can represent D or D' e.g., D-vinyl units, and up to about 10 weight percent vinyl groups based upon Component (A) comprising:

1) $R_3SiO_{1/2}$ units (M or M' units) and SiO_2 units (Q units) where each R is independently selected from the group consisting of vinyl radicals and monovalent hydrocarbon radicals free of aliphatic unsaturation, with a ratio of M units to Q units ranging from about 0.5:1 to about 1.5:1, wherein the copolymer contains from about 0.5 to 10 weight percent vinyl groups, or

2) $R_3SiO_{1/2}$ units (M or M' units), SiO_2 units (Q units) and $R_2SiO_{2/2}$ units (D or D' units) where each R is as defined above and where the ratio of M units to Q units is from about 0.5:1 to about 1.5:1, and the D units are present in the amount from about 1 to 70 mol percent based upon the total number of mols of siloxy units in the copolymer, and where the resinous copolymer contains from about 0.5 to about 10.0 weight percent vinyl groups; or

3) mixtures of units 1 and 2; and

(C) up to about 200 parts by weight of a finely divided silica filler based upon Component (A).

In order to form a heat curable rubber, Component (D) in the form of an organic peroxide free radical initiator or curing agent is provided.

In order to improve processing or properties, the composition may contain one or more of the following:

(E) up to about 10 parts of a processing aid such as a low viscosity silanol stopped siloxane fluid of a about 3 to 500 centipoise at 25°C;

(F) up to about 20 parts by weight of a divinyltetramethyl silazane fluid based upon 100 parts by weight of Component (C); and

(G) up to about 2.5 parts by weight of a cross linking agent such as a polydimethyl siloxane polymethyl hydrogen siloxane random copolymer of about 100 repeating units each of PDMS and PMHS units.

In some applications, (H) a polydiorganosiloxane fluid of about 5,000 to 500,000 cps at 25°C may be employed.

The filler may be treated prior to compounding with up to 20 percent by weight based upon Component (C) of a cyclic methyl tetramer.

The heat curable compositions of this invention have good clarity without sacrificing other beneficial physical properties such as tear strength, tensile strength, hardness, compression set elongation and oil resistance.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a plot illustrating the effect of resin content (Component (B)) on clarity in a clear, heat curable silicone rubber composition of the present invention;

Fig. 2 is a plot illustrating the interaction of the filler treatment and the cross-linking agent on clarity and haze;

Fig. 3 is a plot illustrating the interaction of the filler treatment and water on clarity and haze; and

Fig. 4 is a plot illustrating the interaction of the filler treatment and water on yellowing.

DESCRIPTION OF THE INVENTION

The present invention is directed to a heat curable silicone composition having excellent clarity, good tear and tensile strengths as well as desirable hardness, compression set, elongation and oil resistance. The composition contains a vinyl containing diorgano siloxane gum or gums having a viscosity of about 3,000,000 to 100,000,000 cps at 25°C; an organopolysiloxane resin or resins of the MQ or MDQ types (where each M unit can represent M or M' e.g., M-vinyl units, and where each D unit can represent D or D' e.g., D-vinyl units, and up to about 10 weight percent vinyl groups); and a finely divided silica filler. In accordance with the invention, the resin has a high Q content having sufficient compatibility with the other components so as to provide an essentially one phase system with the gum and filler. Compatibility is defined as a condition where the interface between the components, especially the gum and filler becomes substantially invisible.

Component (A) is a vinyl containing diorgano siloxane gum or blend of such gums having a viscosity from about 3,000,000 to about 100,000,000 cps at 25°C. Preferably, the gum has a viscosity of between about 3,000,000 and 85,000,000 cps, more preferably, about 20,000,000 to about 50,000,000 cps at 25°C. The gum may have a weight percent vinyl concentration of from

about $5x10^{-5}$ to about 1. The gum preferably has a weight percent vinyl concentration in the range from about $6.5x10^{-5}$ to about 0.03, more preferably from about $8x10^{-5}$ to about $1.5x10^{-4}$ and more preferably yet from about $8x10^{-5}$ to about $1.2x10^{-4}$. The organo groups in the vinyl polymer or polymers of the gum should all be monovalent hydrocarbon radicals.

The vinyl polymer or polymers of Component (A) may contain vinyl-on-chain groups and vinyl end-groups. Preferably, the polymer is vinyl terminated. More preferably, the polymer is free of vinyl substitution except at the chain ends. In other preferred embodiments, Component (A) has the formula:

$$ViSiOR^1_2\ (SiOR^1_2)_x(SiOR^2_2)_y\ SiR^1_2Vi$$

wherein Vi is vinyl; R^1 is independently chosen from monovalent hydrocarbon radicals, free of aliphatic unsaturation, and containing 1 to about 8 carbon atoms; R^2 is independently chosen from monovalent hydrocarbon radicals containing 1 to about 8 carbon atoms; and x and y are integers chosen such that Component (A) has a viscosity which ranges from about 3,000,000 to about 85,000,000 cps at 25°C and a weight percent vinyl concentration in the range from about $5x10^{-5}$ to about $2x10^{-4}$, preferably from about $8x10^{-5}$ to about $1.5x10^{-4}$ and more preferably from about $8x10^{-5}$ to about $1.2x10^{-4}$.

The vinyl containing polymers of Component (A) can be made by a process well known in the art, for example, by reacting cyclo-tetrasiloxanes in the presence of low molecular weight, linear vinyl chain stoppers at high temperatures in the presence of basic catalysts so as to yield a polymer of the desired molecular weight. When the reaction is over, the

catalyst is neutralized and the excess cyclics are vented off to result in the desired polymer. By controlling the amount of chain stopper and the temperature of the reaction, the molecular weight of the desired vinyl-containing polymer end product can be controlled. For more details of the process by which such vinyl containing polymers are produced, reference is made, for example, to U.S. Patent No. 3,660,345, which is incorporated herein by reference.

In accordance with the invention, the amount of gum present in the final product may vary. However, for purposes of explanation herein, it is assumed that 100 parts by weight of the gum is combined with varying amounts of the other components. In some examples, the formulation has 80 parts of Component (A). In each case, the amount of other components in the final product may be calculated or otherwise readily inferred. In the examples below Component (A) is a vinyl-stopped polyorganosioxane gum about 9000 units in length.

Component (B) is (1) an organopolysiloxane resin copolymer in an appropriate solvent (e.g. xylene). The resin copolymer may contain $R_3SiO_{1/2}$ monofunctional units (M units) and SiO_2 quadrifunctional units (Q units), where each R is independently selected from the group consisting of vinyl radicals and monovalent hydrocarbon radicals free of aliphatic unsaturation. The ratio of M units to Q units ranges from about 0.5:1 to about 1.5:1, wherein the resin contains about 0.5 to 10.0 weight percent vinyl. Alternatively, Component (B) may comprise (2) an organopolysiloxane resin copolymer containing M and Q units as aforesaid and $R_2SiO_{2/2}$ difunctional (D units) in an appropriate solvent where each R is independently selected from the

group consisting of vinyl radicals and monovalent hydrocarbon radicals free of aliphatic unsaturation. Each M unit may represent M or M′ e.g., M-vinyl units, and each D unit may represent D or D′ e.g., D-vinyl units. The ratio of M units to Q units is from 0.5:1 to about 1.5:1 and the D units are present in an amount of from about 1 to 70 mol percent based upon the total number of mols of siloxy units in the copolymer. The resinous copolymer contains from about 0.5 to about 10.0 weight percent vinyl groups. The organopolysiloxane resin copolymer of Component (B) may contain mixtures of the MQ and the MDQ, resins.

The organopolysiloxane resin copolymers of Component (B) in the present composition and their manufacture are well known in the art. Such resins are usually produced by hydrolysis of chlorosilanes in a process described in U.S. Patent No. 3,436,366 which is incorporated herein by reference.

Component (B) is present in the composition of the present invention in an amount ranging from about 1.0 to about 30 and preferably from 1.0 to about 7.5 parts by weight based upon Component (A). The amount of resin present in the composition, however, is most preferably governed by the amount of clarity desired in the final composition. It has been found that clarity increases greatly when the resin content is between 1 and 7.5 parts by weight relative to 100 parts by weight Component (A). This is best illustrated in Fig. 1 is a graph in which clarity is generally related to weight percent of Component (B) based on Component (A). It can be readily appreciated that the greatest increase in clarity occurs when Component (B) or resin content is between about 1 and about 7.5 parts by weight of Component (A). Thereafter, clarity increases

marginally up to about 25 parts by weight whereupon additional resin concentrations apparently have little or no perceptible positive effect on the clarity.

Component (B′) in the examples below comprises (B2) dispersed in xylene. Much of the xylene has been vacuum stripped so as to result in a composition comprising 60% by weight solids and 40% by weight xylene. The process for manufacturing the composition includes a cook and strip step to facilitate the removal of the residual xylene from the finished compound.

Component (C) comprises from about 5 to about 200, preferably from about 10 to about 100 and more preferably from about 20 to about 75 parts by weight of reinforcing fillers such as SiO_2 based on Component (A). The filler is needed in the composition to provide a high tear strength. Examples of reinforcing fillers that may be used include fumed silica and precipitated silica, with fumed silica being preferred. The filler may be treated with various agents so as to prevent the composition from structuring, for example cyclopolysiloxanes as disclosed in U.S. Patent No. 2,938,009 to Lucas and silazanes as disclosed in U.S. Patent No. 3,635,743 to Smith. The cyclopolysiloxanes present may be, for example, a cyclotetramethyl siloxane present in the amount of about 15 to 20 weight percent of the filler. In the examples below Component (C) is fumed silica treated as in Lucas above having a surface area of about 160 m^2/gm to about 240 m^2/gm. A further surface treatment with a linear vinyl containing silazane may be provided in-situ.

In order to form a heat curable rubber, Component (D), in the form of an organic peroxide free radical initiator or a curing catalyst is provided. The

preferred peroxide curing agents are thermal decomposition organic peroxides conveniently used to cure silicone elastomers. Examples of suitable organic peroxide free radical initiators for use in the present invention are disclosed, for example, in U.S. Patent No. 4,539,357 to Bobear which is incorporated herein by reference. Suitable peroxide catalysts include dialkyl peroxide such as di-tertiary-butyl peroxide, tertiary-butyl-triethylmethyl peroxide, tertiary-butyl-tertiary-butyl-tertiary-triphenyl peroxide, t-butyl perbenzoate and a di-tertiary alkyl peroxide such as dicumyl peroxide. Under certain conditions hereinafter described, such as when a hydride is used, a platinum catalyst may be employed instead of an initiator. In the examples below, the preferred catalyst is a vinyl specific catalyst such as 2,5 dimethyl-2,5-di(t-butyl peroxy) hexane e.g., (Lupersol ™101).

Other components may also be employed as discussed hereinafter. For example, in order to allow for easier incorporation of the filler in the gum, Component (E), a processing aid or plasticizer is employed. In the preferred embodiment, Component (E) is a low viscosity silanol stopped siloxane fluid having a viscosity ranging from about 3-500 cps and preferably 3 to 50 cps at 25°C. The siloxane fluid is an equilibrium mix of low molecular weight oligomers of from about 4 to about 10 but preferably between 4 and 6 repeating units in length with a minimum amount of cyclics in equilibrium with the oligomers. The processing aid (E) may have the form:

$$HO(R_2SiO)_xH$$

where each R is CH_3, and x is between 4 and 10, preferably between 4 and 6, with resulting cyclics of the same number of units in equilibrium.

In the present invention, the processing aid may be present in amounts up to 20 parts by weight based upon Component (A), and preferably is present in amounts between 2 and 10 parts by weight based upon Component (A) and more preferably 3 parts by weight. It should be understood that, typically, the more filler that is used the greater amount of processing aid is employed.

Another Component (F) may be vinyl stopped linear silazane such as divinyltetramethyl silazane. The material may be added for enhancing the bonding of filler to gum. In the present invention, the silazane acts as a filler treatment in-situ. Component (F) is preferably present in an amount up to 2 percent by weight, based upon the weight of Component (A). More preferably, Component (F) is present in an amount of only up to about 0.3 parts by weight, based upon 100 parts by weight of Component (A).

In addition to the foregoing, Component (G) in the form of a hydride cross-linking agent may be employed. In the preferred embodiment, Component (G) may be a random copolymer formed of a polydimethylsiloxane (PDMS) and a polymethylhydrogensiloxane (PMHS) having the form:

$$R_3SiO(SiORHSiOR_2)_xSiR_3$$

wherein each R is independently chosen from a hydrogen or monovalent hydrocarbon radical free of aliphatic unsaturation containing 1 to about 8 carbon atoms, and x ranges from about 2 to about 100. In the present

invention, the hydride is present in an amount of about 0.1 to 10 parts by weight based upon Component (A), preferably 0.5 to 8 parts by weight and more preferably 0.8 to 5 parts by weight based upon Component (A). When a hydride agent is employed, a platinum catalyst may be substituted for the peroxide catalyst to cure the composition. In Component (G) x may vary so that (G) has a viscosity ranging from about 5 to 500 cps, preferably from about 10 to about 100 cps and more preferably from about 10 to 50 cps at 25°C. In the examples below, Component (G) has a viscosity of about 30 centistokes, a hydride content of about 0.8 percent by weight, and a chain length of about 100 units.

The linear hydride described above can be made by many procedures which are known in the art and particularly by the hydrolysis of appropriate chlorosilanes. See for example, U.S. Patent No. 4,041,101 which is incorporated herein by reference.

Component (H) may be present in an amount of up to about 19 parts by weight based upon Component (A). Component (H) is a vinyl-containing diorganopolysiloxane or a vinyl-containing diorganopolysiloxane blend having a viscosity of from about 5000 to about 500,000, preferably from about 40,000 to about 500,000, and most preferably from about 60,000 to about 150,000, centipoise at 25°C.

The vinyl polymer or polymers of Component (H) can be vinyl-terminated and can contain vinyl-on-chain units. In preferred embodiments, however, the vinyl polymer has no vinyl-on-chain units. Preferably, the vinyl polymer of Component (H) has the formula:

$$ViSiOR^1_2(SiOR^2_2)_x(SiOR^2_2)_ySiR^1_2Vi$$

wherein Vi is vinyl, R^1 is selected from the class consisting of vinyl or alkyl radicals of 1 to about 3 carbon atoms, and R^2 is an alkyl radical of 1 to about 8 carbon atoms, x and y vary such that the viscosity of (H) ranges from about 5,000 to about 500,000 centipoise at 25°C, the vinyl concentration being in the range of from about 0.02 to about 0.1 weight percent.

Examples of alkyl radicals which can be represented by R^1 and R^2 include methyl, ethyl, and vinyl radicals. Preferably, R^1 is either vinyl or methyl, and R^2 is methyl.

In order to form a heat curable rubber, an organic peroxide free radical initiator or curing catalyst is provided. The preferred peroxide curing agents are thermal decomposition organic peroxides conveniently used to cure silicone elastomers. Examples of suitable organic peroxide free radical initiators for use in the present invention are disclosed, for example, in U.S. Patent No. 4,539,357 to Bobear which is incorporated herein by reference. Suitable peroxide catalysts include dialkyl peroxide such as di-tertiary-butyl peroxide, tertiary-butyl-triethylmethyl peroxide, tertiary-butyl-tertiary-butyl-tertiary-triphenyl peroxide, t-butyl perbenzoate and a di-tertiary alkyl peroxide such as dicumyl peroxide. Under certain conditions hereinafter described, such as when a hydride is used, a platinum catalyst may be employed instead of an initiator. In the examples below, the preferred catalyst is a vinyl specific catalyst such as 2,5 dimethyl-2,5-di(t-butyl peroxy) hexane e.g., (Lupersol ™ 101).

Other agents may also be added such as small amounts of an acid acceptor, plasticizer, and other materials as long as they do not affect the clarity of

the finished product. The acid acceptor, e.g., MgO soaks up acid liberated during curing. This acid would otherwise cause cleavage of the product chains.

Copending U.S. patent application Serial No. 07/587,876, filed 9/25/90, attorney docket no. 60SI-1336, incorporated herein by reference discloses heat curable silicone rubber compositions containing blends of vinyl containing organopolysiloxane gums and oils, MQ and MDQ resins, filler and a process aid are cured with a peroxide initiator. The compositions are formulated for increased tear strength and reduced compression set properties. While optical properties such as increased clarity and reduced haze are not properties which are discussed in Ward et al. Nevertheless, modified versions of one or more compositions disclosed therein are useful materials for use in a clear, heat curable silicone rubber of the present invention.

In the examples below, all the components were combined in a doughmixer for 4 hours at 160°C. Components (A), (C) and (E) (sometimes hereafter referred to as common components) were compounded with varying amounts of Components (D), (F), (G) and (H).

The common components include:

80 parts by weight of Component (A) comprising a vinyl chainstopped polydimethylsiloxane gum having about 4,000 to about 10,000 repeating units so as to result in a viscosity of about 13 million cps at 25°C;

30 parts by weight of Component (C) comprising a finely divided silica filler having a surface area of about $240m^2/gm$; and

and 4 parts of Component (E) comprising a silanol stopped fluid having a viscosity between 3 and 30 cps at 25°C.

EXAMPLES 1-19

The common components were mixed with various proportions of the following components as follows:

0-10 parts based upon (A) of Component (B') comprising Component (B2) dispersed in xylene as defined above;

0-15 parts of Component (H) comprising a vinyl chain-stopped polydimethylsiloxane high viscosity oil having about 1,000 repeating units and a viscosity of about 60,000 cps at 25 C;

0.6-1.8 parts of Component (G) comprising a polydimethylsiloxane polymethylhydrogensiloxane hydride random copolymer with about 100 repeating units;

0-.15 parts of composition (F) comprising a divinyl tetramethyl silazane; and

0-.1 parts water.

Tables 1A-1C list the resulting compositions and their corresponding properties.

TABLE I - A

EXAMPLE	1	2	3	4	5
Component (Parts by Weight)					
(B') (60 weight percent solids)	5	10	0	10	5
(H)	7.5	0	0	15	7.5
(G)	1.2	1.8	1.8	1.8	1.2
(F)	0.075	0	0.15	0.15	0.075
(H_2O)	0.05	0.1	0.1	0.1	0.05
Properties					
Clarity/Haze	22.1	18.4	27.1	16.6	17.7
Tear "B"	173.0	206.0	224.0	185.0	183.0
Tensile	1827.0	1840.0	1812.0	1642.0	1656.0
Elong. PerCent	925.0	965.0	958.0	807.0	933.0
Hardness Shore A	43.0	48.0	44.0	51.0	46.0
Yellow Index	6.3	6.8	7.2	7.7	6.9
Transmit PerCent	87.6	87.8	85.1	86.1	86.1

TABLE I - B

EXAMPLE	6	7	8	9	10
Component					
(Parts by Weight)					
(B') (60 weight percent solids)	10	5	10	10	0
(H)	15	7.5	0	15	0
(G)	1.8	1.2	0.6	0.6	1.8
(F)	0	0.075	0	0	0
(H_2O)	0	0.05	0	0.1	0
Properties					
Clarity/Haze	16.3	17.0	19.8	17.6	18.6
Tear "B"	155.0	185.0	165.0	125.0	178.0
Tensile	1538.0	1704.0	1483.0	1273.0	1443.0
Elong. PerCent	910.0	936.0	814.0	762.0	1152.0
Hardness Shore A	46.0	44.0	47.0	45.0	35.0
Yellow Index	7.3	6.8	7.2	7.1	7.3
Transmit PerCent	86.5	86.7	86.1	86.4	86.5

TABLE I - C

EXAMPLE	11	12	13	14	15
Component					
(Parts by Weight)					
(B') (60 weight percent solids)	10	0	10	0	10
(H)	0	15	15	15	0
(G)	0.6	0.6	0.6	0.6	1.8
(F)	0.15	0.15	0.15	0	0.15
(H_2O)	0.1	0.1	0	0	0
Properties					
Clarity/Haze	13.3	15.1	15.8	28.4	13.9
Tear "B"	189.0	151.0	164.0	147.0	214.0
Tensile	1537.0	1692.0	1377.0	1297.0	1859.0
Elong. PerCent	667.0	865.0	661.0	971.0	840.0
Hardness Shore A	56.0	40.0	48.0	34.0	58.0
Yellow Index	7.7	8.3	9.0	9.2	6.3
Transmit PerCent	87.0	86.2	85.7	85.2	88.1

Test data were obtained from the following ASTM methods and procedures: Shore A - D-2240; Tensile, Elongation and modulus - D-412 (Die C); Tear - D-624 (Die B); and Compression Set - D-395 (method B).

TABLE I - D

EXAMPLE	16	17	18	19
Component (Parts by Weight)				
(B') (60 weight percent solids)	0	0	0	0
(H)	15	0	0	15
(G)	1.8	0.6	0.6	1.8
(F)	0.15	0	0.15	0
(H_2O)	0	0.1	0	0.1
Properties				
Clarity/Haze	15.3	22.1	18.7	23.5
Tear "B"	164.0	179.0	206.0	136.0
Tensile	1746.0	1228.0	1727.0	1584.0
Elong. PerCent	954.0	975.0	898.0	1104.0
Hardness Shore A	41.0	36.0	43.0	30.0
Yellow Index	7.4	8.6	9.4	8.1
Transmit PerCent	85.9	85.1	85.2	84.5

In each of the Tables IA, IB, IC and ID the common components were combined with the various proportions of the components as noted to make up the individual examples.

All of the results for the properties are to the fourth decimal place.

TABLE II

Composition	Haze %	Yellow Index	Trans %	Tear B pi	Shore A	Tensile PSI	Elong %
Resin (B') (60 wt % solids)	-	-	+		+		-
Vinyl Terminated oil (H)				-	-	-	
Hydride (G)		-		+		+	+
LVS (F)	-	-		+	+	+	-
H_2O							
Interacting Compositions							
(G)/H_2O	S	S		S			
(G)(F)	S						
(B')/(G)				S			
(B')/(H)						S	
(B')/(F)						S	

+	Increases Number Value of Property
-	Reduces Number Value of Property
S	Significant effect
(blank)	no significant effect

Table II represents a tabulation of results observed when various properties were evaluated for EXAMPLES 1-19. The upper half of the table lists the five modifiers and their particular effect on the various properties. As set forth in the table, a plus sign (+) indicates that the particular modifier has a positive or increasing effect on the numerical value of the measured property and a minus sign (-) indicates that the modifier decreases the numerical value of the measured property. S indicates significant effects are measured; and a blank indicates no significant effects are measured. In accordance with the invention, it can be appreciated that the amount of resin, Component (B'), has a positive effect on transmission or clarity increasing the % transmission and decreasing both the haze and yellow index. Component (H) is normally used as a resin carrier and has no significant effect on the clarity but may suppress tear B, Shore A, and tensile strength. The hydride cross linking agent, Component (G) decreases yellowness and has a positive effect on strength. Likewise increasing amounts of linear vinyl silazane (F) decreases haze and yellowness, has a negative effect on elongation and has a positive effect on tear B, Shore and tensile strength. The water does not seem to have any significant effect on the properties directly.

The lower half of Table II illustrates the interactions between the components. It can be seen that the hydride, Component (G) and the water have significant interacting effects on haze, yellowness and tear B. Likewise the hydride and linear vinyl silazane (F) have a significant effect on haze. The resin (B') interacts with the hydride (G) to effect tear B and the

resin (B') interacts with both the oil (G) and the linear vinyl silazane (F) to effect tensile strength.

Fig. 2 illustrates the interaction between the silazane (F) and the hydride (G). The plot illustrates that increasing amounts of the silazane decreases the haze and yellowness the most when the hydride is reduced.

Fig. 3 shows that haze is reduced and therefore clarity increases when the water content is high with a corresponding reduced hydride (G) content or with a reduced water content and increasing hydride content.

Fig. 4 graphically illustrates that yellowness is decreased with increasing hydride content. However, increasing the water content suppresses the advantageous reduction in yellowness.

Tables III-(A), (B) and (C) numerically illustrate the effects graphically illustrated in Figs. 2-4. When the hydride (G) and silazane (F) are compared, haze is reduced when the silazane content is High and the hydride content is Low, which is desirable in accordance with the present invention. Likewise when the water content is reduced in comparison to the hydride, haze is reduced (clarity is improved). Finally, the yellowness is reduced as the hydride content increases with decreasing water content, as illustrated on Table III-(C).

Minimum and maximum values for the various properties are also set forth in Tables III-(A),(B) and (C). However, from the foregoing it should be readily apparent that the most pronounced improvement in the clarity results by increasing the amount of resin in the composition so as to best match the refractive indices of the gum (Component A) and the filler (Component C).

TABLE III - (A)

Components (F) - (G)	Haze
Low - Low	21.97
High - Low	15.72
Low - High	19.20
High - High	18.22
Maximum	28.40
Minimum	13.30

TABLE III - (B)

Components H_2O - (G)	Haze
Low - Low	20.67
High - Low	17.02
Low - High	16.02
High - High	21.40
Maximum	28.40
Minimum	13.30

TABLE III - (C)

Components (G) - H_2O	Yellowness
Low - Low	8.700
High - Low	7.075
Low - High	7.925
High - High	7.450
Maximum	9.400
Minimum	5.300

Although the present invention has been described with reference to specific embodiments, it is to be understood that substitutions, additions, deletions and modifications within the skill of an ordinary artisan can be made without departing from the scope of the invention as defined in the appended claims.

WHAT IS CLAIMED IS:

1. A clear heat cured silicone rubber composition comprising in parts by weight:

(A) 100 parts by weight of a vinyl containing organopolysiloxane having a viscosity of about 3,000,000 to about 100,000,000 cps at 25°C;

(B) about 1 to about 30 parts by weight based upon Component (A) of an organopolysiloxane resin copolymer comprising

1) $R_3SiO_{1/2}$ units (M or M′ units) and SiO_2 units (Q units) where each R is independently selected from the group consisting of vinyl radicals and monovalent hydrocarbon radicals free of aliphatic unsaturation with a ratio of M units to Q units ranging from 0.5:1 to about 1.5:1, where the copolymer contains from about 0.5 to 10 weight percent vinyl groups, or

2) $R_3SiO_{1/2}$ units (M or M′ units), SiO_2 units (Q units) and $R_2SiO_{2/2}$ units (D or D′ units) where each R is as defined above and where the ratio of M or M′ units to Q units is from 0.5:1 to about 1.5:1, and the D or D′ units are present up to about 70 mol percent based upon the total number of mols of siloxy units in the copolymer, and where the resinous copolymer contains from about 0.5 to about 10.0 weight percent vinyl groups, or

3) mixtures of 1 and 2; and

(C) up to about 200 parts by weight of a finely divided silica filler based upon Component (A).

2. The clear, heat cured rubber of claim 1, further comprising (D) an organic peroxide free radical initiator or curing agent.

3. The clear, heat cured rubber of claim 1, further comprising:

(E) up to about 10 parts by weight of a low viscosity silanol stopped siloxane fluid of about 3 to about 500 cps at 25°C based upon Component (A).

4. The clear, heat cured rubber of claim 1, further comprising Component (F) up to about 3 parts by weight of a divinyltetramethyl silazane fluid based upon 100 parts of Component (A).

5. The clear, heat cured rubber of claim 1, further comprising Component (G) up to about 2.5 parts by weight of a polydimethylsiloxane (PDMS) polymethylhydrogensiloxane (PMHS) random copolymer of about 100 repeating units each of PDMS and PMHS based upon Component (A).

6. The clear, heat cured rubber of claim 1, wherein said filler, Component (C), comprises fumed silica treated with up to 20 percent by weight of said filler of a cyclic methyl tetramer.

7. The clear, heat cured rubber of claim 1, further comprising Component (H) up to about 19 parts by weight of a polydiorganosiloxane fluid of about 5,000 to 500,000 cps at 25°C based upon Component (A).

8. The clear, heat cured rubber of claim 1, comprising about 20 to about 75 parts by weight of Component (C) based upon Component (A).

9. The clear, heat cured rubber of claim 1, wherein Component (B2) is present in an amount of about

1 to about 7.5 parts by weight based upon Component (A).

10. The clear, heat cured rubber of claim 1, wherein Component (B) is dispersed in xylene and vacuum stripped so as to result in a composition comprising 60% by weight solids and 40% by weight xylene.

11. The clear, heat cured rubber of claim 10, where residual xylene has been removed from the finished compound.

12. The clear, heat cured rubber of claim 1, wherein Component (B) has a Q content in an amount sufficient to result in compatibility between Components (A) and (C) so as to form an essentially one phase system of Components (A), (B) and (C).

13. The clear, heat cured rubber of claim 12, where compatibility is defined as a condition where the interface between the components in the composition is essentially invisible.

14. The clear, heat cured rubber of claim 1, having a transmission of at least about 84%.

15. The clear, heat cured rubber of claim 1, having a yellow index less than about 9.5%.

16. The clear, heat cured rubber of claim 1, having a yellow index of about 6.3% to about 9.5%.

17. The clear, heat cured rubber of claim 1, having a compression set of less than about 25.5%.

18. The clear, heat cured rubber of claim 1, having a compression set of less than about 20%.

19. The clear, heat cured rubber of claim 1, having a hardness of at least about 30 on a Shore A scale.

20. The clear, heat cured rubber of claim 1, having a hardness of between about 30 and 58 on a Shore A scale.

21. The clear, heat cured rubber of claim 1, having an elongation of at least 677%.

22. The clear, heat cured rubber of claim 1, having an elongation of between about 650% and 1,150%.

23. The clear, heat cured rubber of claim 1, having a tensile strength of at least about 1,200 PSI.

24. The clear, heat cured rubber of claim 1, having a tensile strength of between about 1,200 PSI and about 1,860 PSI.

25. The clear, heat cured rubber of claim 1, having a tear B of at least 125 pi.

26. The clear, heat cured rubber of claim 1, having a tear B of between about 125 to about 224 pi.

27. The clear, heat cured rubber of claim 1, having a haze of less than about 24%.

28. The clear, heat cured rubber of claim 1, having an haze between about 13% and about 24%.

29. The clear, heat cured rubber of claim 1, wherein the filler has a surface area of at least 100 square meters per gram.

30. The clear, heat cured rubber of claim 1, wherein the filler has a surface area of about between 100 and 300 square meters per gram.

31. The clear, heat cured rubber of claim 30, wherein the filler has a surface area of between about 160 and 240 square meters per gram.

32. The clear, heat cured rubber of claim 1, wherein Component (A) as the structure

$$ViSiOR^1_2\ (SiOR^1_2)_x(SiOR^2_2)_y\ SiR^1_2Vi;$$

wherein Vi is vinyl and each R1 is independently chosen from monovalent hydrocarbon free radicals free of aliphatic unsaturation and containing 1 to about 8 carbon atoms;

each R^2 is independently chosen from monovalent hydrocarbon radicals containing 1 to about 8 carbon atoms, and x and y are integers chosen such that the viscosity is about 3M to about 100 M cps at 25°.

33. The clear, heat cured rubber of claim 32, wherein the vinyl concentration weight percent is in range from about $5x10^{-5}$ to about 1.

34. The clear, heat cured rubber of claim 32, wherein the vinyl concentration is in a range of about $6.5x10^{-5}$ to about 0.03.

35. The clear, heat cured rubber of claim 32, wherein the vinyl concentration is in a range of about $5x10^{-5}$ to about $2x10^{-4}$.

36. The clear, heat cured rubber of claim 32, wherein the vinyl concentration is in a range from about $8x10^{-5}$ to about $1.5x10^{-4}$.

37. The clear, heat cured rubber of claim 32, wherein the vinyl concentration is in a range of about $8x10^{-5}$ to about $1.2x10^{-4}$.

Fig. 1.

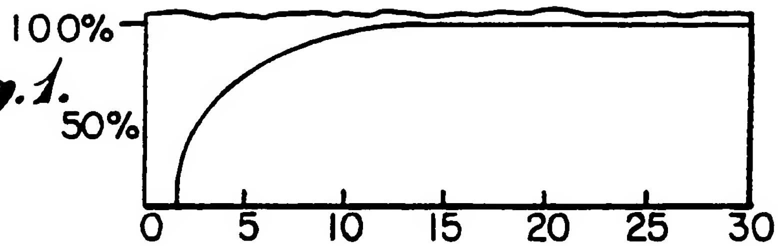

Fig. 2.

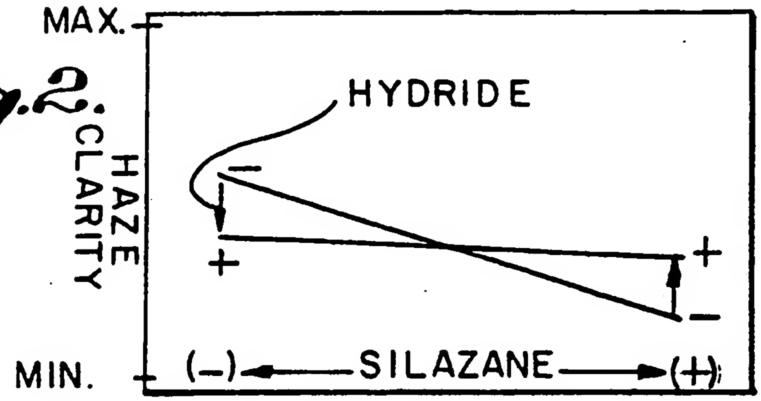

Fig. 3.

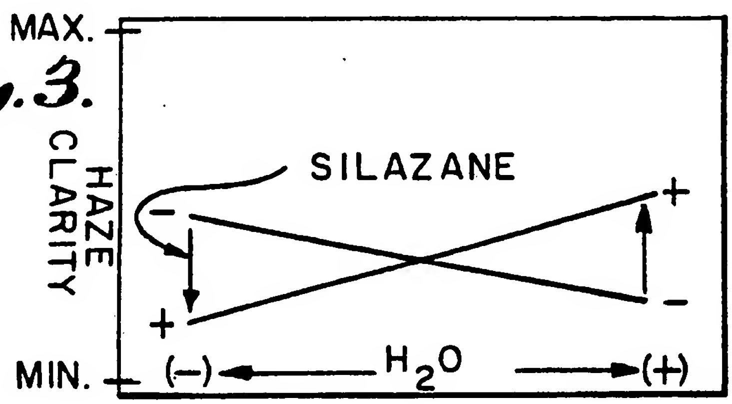

Fig. 4.